Imaging Detectors Based on the Response of Anisotropic Layered Materials

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Abstract—**The inherent properties of naturally layered materials such as the high-temperature superconductors cause a transient voltage when a light pulse is absorbed. The amplitude is proportional to the absorbed energy. Observable pulses arise in the HTS materials even at room temperature. A phenomenological model of the effects consistent with our observations on the first generation of sensors will be discussed along with recent results regarding sensitivity to single photons, array geometry and readout, and energy resolution.**

I. Introduction

Non-dispersive methods of determining the single particle/quantum energy are of interest for both space research and laboratory instrumentation.

The *active hot-electron devices* [1,2] have demonstrated advantages over earlier designs and useful energy resolution in single pixel designs.

The concept of a *passive hot-electron sensor* has been developed by our group [3]. An unbiased pixel transforms the captured energy into the voltage using thermoelectric devices. The analysis suggests that sub-eV energy resolution of single high-energy quanta may be possible at MHz rates in a multi-pixel array. An important option of these detectors utilizes anisotropic thermoelectric sensors. Three independent development directions in solid state physics led to this option. One direction is the development of novel oxide-layered materials, which reveal anomalously high voltage response to laser radiation [4]-[11]. The second direction was initiated by the Chernovtsy group (see review article [12]) and involves a theoretical understanding of anisotropic thermoelectricity. The third direction came from our research into the non-equilibrium μ-potential in layered oxides as a basis for single-particle detection: the signals measured by our group [13] were at least in part caused by the same anisotropic thermoelectricity.

This report presents a thorough critical analysis of anisotropic thermoelectricity as applied to radiation detectors.

II. Physics of the Sensor

Oxide superconductors have an inherently layered structure. This causes their transport properties within and between the layers to be very different – the anisotropy in the normal state electric conductivity can be 10^4 [14]. Within the crystal unit cell, the high conductivity planes are referred to as the ab-planes, while the perpendicular direction is called the c-axis. Deposition technology exists to grow these materials with a uniform crystallographic orientation on special (lattice matched) substrates so that the films resemble thin single crystals. Consider what happens when the top surface of the film is ΔT warmer than the bottom of the film (Fig. 1). If the material has **isotropic** properties and the temperature gradient is time independent, a DC voltage will be established across the thickness of the film (in **z**-direction, **parallel**

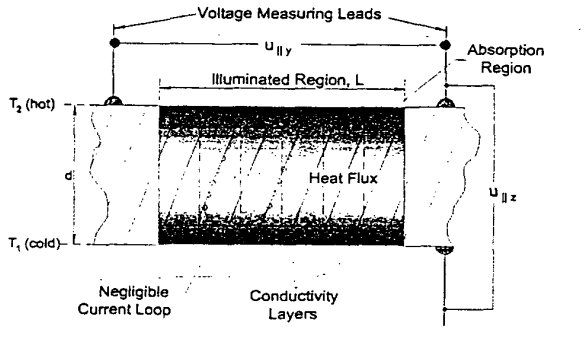

Fig. 1. Surface heating by laser illumination produces vertical thermal gradient and transverse voltage. The high anisotropy suppresses electron transport between layers and inhibits circulating currents.

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to the temperature gradient):

$$V = \varphi_2 - \varphi_1 = S\,\Delta T, \quad (1)$$

where S is the usual Seebeck coefficient and $\Delta T = T_2 - T_1$.

In **anisotropic** materials the Seebeck coefficient is not a scalar, but rather becomes a tensor: $S \rightarrow S_{ik}$. Then:

$$\nabla_k \varphi = \Sigma_m S_{km} \nabla_m T. \quad (2)$$

In the situation depicted in Fig.1, symmetry will cause the heat flow to be directed perpendicular to the surface of the film (**z**-direction), so that the m =3 (i.e., z) component of ∇T in Eq. (2) is non-zero.

In the crystallographic reference frame X′, tilted in the (**y**-**z**)-plane by angle α, the tensor S'_{km} is diagonal: S'_{yy}, $S'_{zz} \neq 0$, and $S'_{yz} = S'_{zy} = 0$. Because the **x** axis is unchanged between the two reference frames and there is no thermal gradient in that direction,

$$V_{\|x} = 0. \quad (3)$$

In the measurement frame of reference, the non-diagonal components of the Seebeck tensor also are non-zero. As follows from (2), the non-zero value of $(\nabla T)_z$ becomes coupled with the S_{yz} and S_{zz} components. The measurable potential difference along the direction of temperature gradient is of the usual sort for thermoelectricity:

$$V_{\|z} = \{\cos 2\alpha (S'_{zz} - S'_{yy}) + (S'_{zz} + S'_{yy})\}(T_1 - T_2)/2 \equiv S_{\|z}^{\text{eff}} \Delta T. \quad (4)$$

Assuming the $(\nabla T)_z$ is constant along the y direction, we get

$$V_{\|y} = (S'_{yy} - S'_{zz}) \sin 2\alpha (\nabla T)_z L/2 \equiv S_{\|y}^{\text{eff}} \Delta T (L/d). \quad (5)$$

Equation (5) says that, because of the layered anisotropic structure, a **vertical temperature gradient produces a lateral voltage**. Moreover, $V_{\|y}$ is much larger than $V_{\|z}$ assuming $(L/d) >> 1$.

III. Experimental Results

There are a number of qualitative and quantitative consequences of this theoretical model which have been experimentally confirmed by our group. In our measurements, YBCO films, biased in the normal state, were heated using laser-pulses.

Equations (3) and (5) predict that the measured voltage should strongly depend on the angular relationship of the line between the voltage probes and the projection of the crystallographic c axis. The patterning shown in Fig. 2 was made to investigate this predicted dependence, which was confirmed.

To test the dependence in Eq. (5) on the tilt angle α of the c axis of the material, YBCO films were deposited on special vicinally cut substrates to produce c axis inclinations of 0.03°, 5° and 20° as measured by 4 circle x-ray

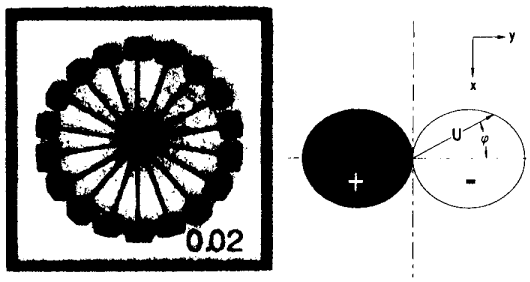


Fig. 2. Photograph of the film patterned to reveal the dependence of the voltage on the orientation. The response plotted in polar coordinates.

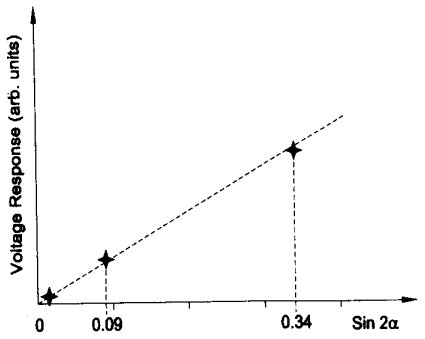
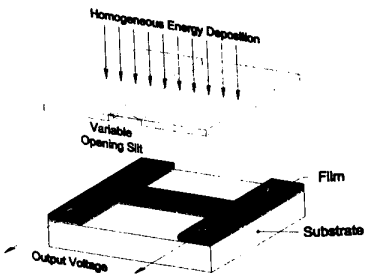


Fig. 3. Dependence of $V_{\|y}$ on the inclination angle α, Eq. (5).



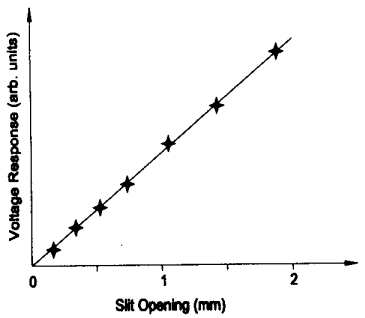


Fig. 4. Strip geometry sample illuminated through a variable width slit: linear dependence on slit opening (parameter L in Eq. (5)).

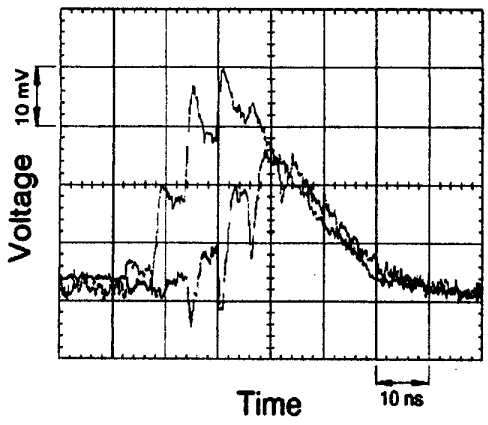


Fig. 5. Signal registered when pulsed UV radiation is incident upon the front side of the sensor (upper curve), and upon the backside of the film after passing through the substrate (lower curve).

diffractometer. The result shown in Fig. 3 demonstrates the expected linear dependence of maximum output voltage on sin2α. Equation (5) also asserts that the voltage observed should depend on the length of the heated area L. Illuminating a strip-like sample homogeneously through a slit with a variable opening, the expected linear dependence results (Fig. 4). The amplitude of the voltage signal is independent of the extent of the sensor (width) along the **a** axis of the material(**x** axis in the measurement frame of reference).

The proportionality of the signal to ∇T was confirmed both by increasing the deposited energy and by decreasing the thickness d of the sensor film.

Sub-ns laser pulses and fast signal acquisition reveal both fast and slow components of the signal (see Fig. 5). The fast signals change sign when the illumination is changed from the front to back side of the transparent substrate (cf. curves a and b in Fig. 5). This makes it clear that fast signals are related to heat fluxes within the film. The slow component has the same sign at both types of energy deposition. Thus it is related to the heat flow out of the film, into the substrate.

The amplitude of the signal agrees well with the prediction of Eq. (5): at 300K, a raise of temperature estimated as ~ 0.5–1 K produces a voltage pulse 10 mV in amplitude in a sample with parameters $\alpha = 5°$, d = 500 nm, and L= 2 mm. This means the effective Seebeck coefficient $(S'_{yy} - S'_{zz})$ entering Eq. (5) has a value ~10 μV/K which is quite consistent with literature values of the thermoelectricity in YBCO.

IV. Thermal Modeling of the Detector

A passive hot-electron microcalorimeter would be constructed with a distinct absorber volume V_{abs} that has a thickness optimized to stop the requisite energy photons.

The operating temperature T should be low in order to reduce the thermal capacity of the absorber and thermal fluctuations, typically T~0.1K.

The time dependent temperature excursion $\delta T = T^* - T$ has its maximum value Q/C_{abs}, where C_{abs} is the heat capacity of the absorber, and Q is deposited energy. The heat-sink (substrate) stays at T. The heat diffuses from the absorber to the heat sink via the thermoelement. This energy flux creates the voltage signal that lasts approximately

$$\tau = C_{abs}/G, \tag{6}$$

where G is the thermal conductance via thermoelement and its interfaces with the absorber and the substrate.

Since the absorber is a normal-metal film of a restricted geometry, with typical size ~ 10 μm, the electron temperature homogenization in it is a fast process, usually being considered as "instantaneous". In the next stage, the excess energy is transferred to the sensor film through an insulating barrier. The primary function of the barrier is to prevent the metallic absorber from shorting out the signal in the sensor underneath. Another, very useful function, is the heat transfer regulation.

We have analyzed the hot electron transport through the barrier in two different models. In the first one, the insulating barrier is considered as a layer of very high impurity concentration metal. Elastic tunneling is the opposite considered case of hot-electron penetration across the insulator. In both cases the time τ at which the electrons propagate from the metallic absorber into the sensor is in the microsecond range, provided the parameter RA is about 10^{-6} Ω-cm^2 (A is the absorber cross-section, R is the barrier resistance) and the absorber is ~1 μm thick metal. Also, since $\tau \propto (RA)^{-1}$, by making RA ~ 10^{-5} Ω-cm^2, one can get τ ~ 10 μs, which will match better the time of the energy outflow through the Kapitza boundary between sensor and dielectric substrate.

For even so long a time as 10 μs-duration time-scale the main role in heat conduction may be played by hot electrons, while the phonons are inefficient. This could be produced either by lowering the bias temperature (so that the rate of electron intercollisions $\gamma_{e\text{-}e} \propto T^2/\varepsilon_F$ becomes higher than the rate of electron-phonon collisions $\gamma_{e\text{-}ph} \propto T^3/\Omega_D^2$) and/or by the particular choice of absorber material (in transition metals, such as tungsten, even at T≥4K, $\gamma_{e\text{-}e} >> \gamma_{e\text{-}ph}$ [15]).

Of course, if the signal can be time-integrated, the duration of the pulse is not as important. Indeed, in our vertical stack geometry, heat has no way to go other than through the sensitive element. In this case one can evoke the Fourier law to couple the ∇T with the value of $\mathbf{J}_H$, the energy ("heat") flux through the sensor

$$\mathbf{J}_H = -k_{eff} \nabla T, \tag{7}$$

Here k_{eff} is regarded as an effective (electron, phonon, etc.) heat conductivity of the sensor. Consider an arbitrary cross-section of the sensor perpendicular to the **z** direction. Then

the rate of energy flow from the absorber through this area $A=L^2$ at any given time will be

$$dQ/dt = \int dA \cdot J_H = -A\, k_{eff} (\nabla T)_z. \quad (8)$$

Using (7), (8) and (5), one can find:

$$\int V(t)\, dt \equiv V_{signal} \times \tau_{signal} = \{S_{eff}(L/d)\}\, (Q/G). \quad (9)$$

The important conclusion is that this integral quantity is independent on C_{abs}. It is also proportional to the value of deposited energy Q, which permits to use the device as the energy sensor.

For the heat conductance of the sensor layer it follows

$$G = k_{\|z}\, A/d, \quad (10)$$

where $k_{\|z}$ is the heat conductivity along z-direction. Applying the Wiedemann-Frantz law, we have:

$$V_{signal} \times \tau_{signal} = (dS_{eff}/dT) R_{\|z}\, (L/d)\, Q/L_0 \quad (11)$$

(L_0=25 nWΩ/K^2). Let us assume that the input impedance of the external circuit is much smaller than $R_{\|y}$, as will be the case if a superconducting read-out circuit is used. Then the output of the sensor could be characterized in terms of an effective charge:

$$q_{signal} = \int I_{signal} dt = (R_{\|z} / R_{\|y})\, (dS_{eff}/dT)\, (L/d)\, Q/L_0. \quad (12)$$

Eq. (12) will now be used to optimize the performance of the detector.

V. Optimal Signal Acquisition

In the isotropic case [3] we have just

$$q_{signal} = (dS/dT)\, Q/L_0, \quad (13)$$

and for dS/dT~30 μV/K^2, $q_{signal}=10^3$ electrons at Q~1eV. We should mention that both the factor $(R_{\|z}/R_{\|y})$ and the function S_{eff} are dependent on the film tilt angle, α. Optimization with respect to α of the product of these factors gives:

$$q_{signal} = (\sigma'_{\|y}/\sigma'_{\|z})^{1/2}\, (d/L)\{[d(S'_{yy} - S'_{zz})]/dT\}\, Q/L_0 \quad (14)$$

(this corresponds to the optimal α being $\sim(\sigma'_{\|z}/\sigma'_{\|y})^{1/2}$).

As was mentioned, the factor $\sigma'_{\|y}/\sigma'_{\|z}$ could be as large as 10^4, so for a sensor with the aspect ratio L/d=20 one can have a factor of 5 higher q_{signal} than with the isotropic case, provided the temperature derivatives of the Seebeck coefficients in (13) and (14) are the same.

Semiconductor-based amplifiers exist with room temperatures noise levels of ~100 electrons, and SQUID-based amplifiers at sub-K temperatures should have much lower noise. Thus the signal-to noise ratio should be set by the noise of the sensor. This is expected to be dominated by the inherent Johnson noise of the resistor $R_{\|y}$. For a circuit with electric capacitance C_{el} and operational bandwidth $\delta\nu_B$, the charge equivalent noise is

$$q_{noise} = C_{el} \{4 k_B T_{op} R_{\|y}\, \delta\nu_B\}^{1/2}. \quad (14)$$

At T_{op}=0.1K, $R_{\|y}$ =10Ω and $\delta\nu_B$~1MHz (consistent with 1 μs events), we will have $q_{noise}\approx$10 electrons at C_{el} ~ 10 pF. This noise level is higher than in the case of isotropic sensors [3], but still permits resolution approaching 1eV if a layered material with the desired values of dS/dT can be identified.

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